

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

Attorney's Docket Number
05725.0398

U.S. Application No.

09/319204

International Application. No.	International Filing Date	Priority Date Claimed
PCT/FR98/02074	September 28, 1998	October 3, 1997

Title of Invention:

COMPOSITION FOR THE OXIDATION DYEING OF KERATIN FIBRES AND DYEING PROCESS USING THIS COMPOSITION

Applicants For DO/EO/US:

Roland DE LA METTRIE, Jean COTTERET, Arnaud DE LABBEY, and Mireille MAUBRU

Applicants herewith submit to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☐ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☒ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes (Amended Sheets) to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).
11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☐ A FIRST preliminary amendment.
14. ☐ A SECOND or SUBSEQUENT preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ Other items or information:
 - a. ☐ Verified Small Entity Statement.
 - b. ☐ Annexes (Amended Sheets) to Intl. Preliminary Examination report.

17. [X] The following fees are submitted:

CALCULATIONS

Basic National Fee (37 CFR 1.492(a)(1)-(5)):

Search Report has been prepared by the EPO or JPO.....\$840.00
 International preliminary examination fee paid to
 USPTO (37 CFR 1.482).....\$670.00
 No international preliminary examination fee paid to
 USPTO (37 CFR 1.482) but international search fee
 paid to USPTO (37 CFR 1.445(a)(2)).....\$760.00
 Neither international preliminary examination fee
 (37 CFR 1.482) nor international search fee
 (37 CFR 1.445(a)(2)) paid to USPTO.....\$970.00
 International preliminary examination fee paid to USPTO
 (37 CFR 1.482) and all claims satisfied provisions
 of PCT Article 33(1)-(4).....\$ 96.00

ENTER APPROPRIATE BASIC FEE AMOUNT = \$ 840.00

Surcharge of \$130.00 for furnishing the oath or declaration later than
 [] 20 [] 30 months from the earliest claimed priority date
 (37 CFR 1.492(e)). \$

Claims	Number Filed	Number Extra	Rate	
Total Claims	33-20=	13	X \$18.00	\$ 234.00
Independent Claims	1- 3=		X \$78.00	\$
Multiple dependent claim(s) (if applicable)			+\$260.00	\$ 260.00

TOTAL OF ABOVE CALCULATIONS = \$1334.00

Reduction by 1/2 for filing by small entity, if applicable. Verified
 Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28) \$

SUBTOTAL = \$1334.00

Processing fee of \$130.00 for furnishing the English translation later
 than [] 20 [] 30 months from the earliest claimed priority date
 (37 CFR 1.492(f)). + \$

TOTAL NATIONAL FEE = \$1334.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The
 assignment must be accompanied by an appropriate cover sheet
 (37 CFR 3.28, 3.31). \$40.00 per property + \$

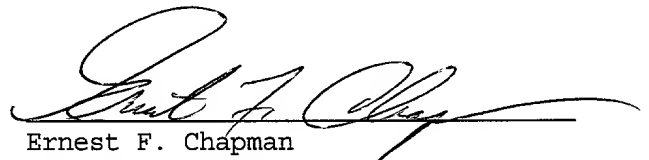
TOTAL FEES ENCLOSED = \$1334.00

Amount to be
 refunded \$
 charged \$

- a. [X] A check in the amount of **\$1334.00** to cover the above fees is enclosed.
 b. [] Please charge my Deposit Account No. _____ in the amount of \$ _____
 to cover the above fees. A duplicate copy of this sheet is enclosed.
 c. [X] The Commissioner is hereby authorized to charge any additional fees
 which may be required, or credit any overpayment to Deposit Account
 No. 06-0916. A duplicate copy of this sheet is enclosed.

The Commissioner is hereby authorized to charge any other fees due under 37 C.F.R. §1.16
 or §1.17 during the pendency of this application to our Deposit Account No. 06-0916.

SEND ALL CORRESPONDENCE TO:
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 Ernest F. Chapman
 Reg. No. 25,961

05725.0398

Submitted: June 02, 1999

PATENT

Attorney Docket No. 05725.0398-00

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re National Stage of International
Application No. PCT/FR98/02074

Roland DE LA METTRIE et al.

Serial No.: 09/319,204

PCT Filed: September 28, 1998

National Stage Entry: June 2, 1999

For: COMPOSITION FOR THE OXIDATION
DYEING OF KERATIN FIBRES AND
DYEING PROCESS USING THIS
COMPOSITION

Group Art Unit: Unassigned

Examiner: Unassigned

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

Prior to the examination of this application, please amend the specification
and claims as follows:

IN THE CLAIMS:

Please cancel claims 1 to 31, without prejudice or disclaimer, and replace
them with new claims 32 to 74 as follows:

--32. A ready-to-use composition for the oxidation dyeing of keratin fibers,
comprising:

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36. The composition according to Claim 32, wherein said at least one 2-electron oxidoreductase is chosen from uricases of animal, microbiological and biotechnological origin.

37. The composition according to Claim 32, wherein said at least one 2-electron oxidoreductase is present in an amount ranging from 0.01 to 20% by weight relative to the total weight of the composition.

38. The composition according to Claim 37, wherein said at least one 2-electron oxidoreductase is present in an amount ranging from 0.1 to 5% by weight relative to the total weight of the composition.

39. The composition according to claim 32, wherein said at least one donor for said at least one 2-electron oxidoreductase is chosen from D-glucose, L-sorbose, D-xylose, glycerol, dihydroxyacetone, lactic acid and its salts, pyruvic acid and its salts, and uric acid and its salts.

40. The composition according to Claim 39, wherein said at least one donor for said at least one 2-electron oxidoreductase is chosen from uric acid and its salts.

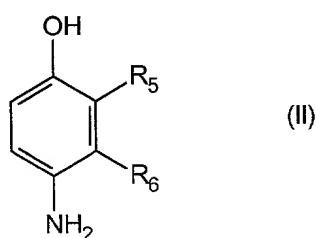
41. The composition according to Claim 32, wherein said at least one donor is present in an amount ranging from 0.01 to 20% by weight relative to the total weight of the composition.

42. The composition according to Claim 41, wherein said at least one donor is present in an amount ranging from 0.1 to 5% by weight relative to the total weight of the composition.

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43. The composition according to Claim 32, wherein said para-aminophenols are chosen from compounds corresponding to formula (II) below, and acid-addition salts thereof:



in which:

- R_5 is chosen from a hydrogen atom, halogen atoms, C_1 - C_4 alkyl radicals, C_1 - C_4 monohydroxyalkyl radicals, $(C_1$ - C_4)alkoxy $(C_1$ - C_4)alkyl radicals, C_1 - C_4 aminoalkyl radicals, and hydroxy $(C_1$ - C_4)alkylamino $(C_1$ - C_4)alkyl radicals,
 - R_6 is chosen from a hydrogen atom, halogen atoms, C_1 - C_4 alkyl radicals, C_1 - C_4 monohydroxyalkyl radicals, C_2 - C_4 polyhydroxyalkyl radicals, C_1 - C_4 aminoalkyl radicals, cyano $(C_1$ - C_4)alkyl radicals, and $(C_1$ - C_4)alkoxy $(C_1$ - C_4)alkyl radicals,
- wherein at least one of the radicals R_5 and R_6 represents a hydrogen atom.

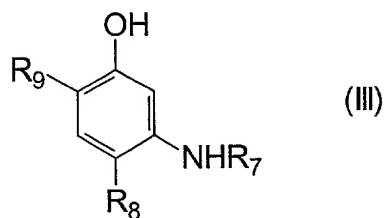
44. The composition according to Claim 43, wherein said para-aminophenols of formula (II) are chosen from para-aminophenol, 4-amino-3-methylphenol, 4-amino-3-fluorophenol, 4-amino-3-hydroxymethylphenol, 4-amino-2-

methylphenol, 4-amino-2-hydroxymethylphenol, 4-amino-2-methoxymethylphenol, 4-amino-2-aminomethylphenol, 4-amino-2-(β -hydroxyethylaminomethyl)phenol, 4-amino-2-fluorophenol, and acid addition salts thereof.

45. The composition according to Claim 32, wherein said at least one second oxidation base is chosen from para-aminophenols present in an amount ranging from 0.0005 to 12% by weight relative to the total weight of the composition.

46. The composition according to Claim 45, wherein said at least one second oxidation base is chosen from para-aminophenols present in an amount ranging from 0.005 to 6% by weight relative to the total weight of the composition.

47. The composition according to Claim 32, wherein said at least one coupler is chosen from meta-amino phenols of formula (III) below, and acid-addition salts thereof:



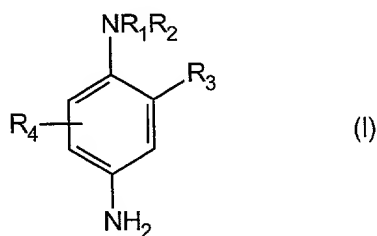
in which:

48. The composition according to Claim 47, wherein said at least one coupler of formula (III) is chosen from meta-aminophenol, 5-amino-2-methoxyphenol, 5-amino-2-(β -hydroxyethyloxy)phenol, 5-amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-4-methoxy-2-methylphenol, 5-amino-4-methoxy-2-methylphenol, 5-amino-4-chloro-2-methylphenol, 5-amino-2,4-dimethoxyphenol, 5-(γ -hydroxypropylamino)-2-methylphenol and acid-addition salts thereof.

50. The composition according to Claim 49, wherein said at least one coupler is present in an amount ranging from 0.005 to 5% by weight relative to the total weight of the composition.

51. The composition according to Claim 47, wherein said halogen atoms are chosen from chlorine, bromine, and fluorine.

52. The composition according to Claim 32, wherein said para-phenylenediamine compounds are chosen from compounds of formula (I) below, and the acid-addition salts thereof:



in which:

- R_1 is chosen from a hydrogen atom, C_1 - C_4 alkyl radicals, C_1 - C_4 monohydroxyalkyl radicals, C_2 - C_4 polyhydroxyalkyl radicals, $(C_1$ - C_4)alkoxy $(C_1$ - C_4)alkyl radicals, C_1 - C_4 alkyl radicals substituted with a nitrogenous group, a phenyl radical and a 4'-aminophenyl radical;
- R_2 is chosen from a hydrogen atom, C_1 - C_4 alkyl radicals, C_1 - C_4 monohydroxyalkyl radicals, C_2 - C_4 polyhydroxyalkyl radicals, $(C_1$ - C_4)alkoxy $(C_1$ - C_4)alkyl radicals and C_1 - C_4 alkyl radicals substituted with a nitrogenous group;

- R₃ is chosen from a hydrogen atom, halogen atoms, C₁-C₄ alkyl radicals, C₁-C₄ monohydroxyalkyl radicals, C₁-C₄ hydroxyalkoxy radicals, acetamino(C₁-C₄)-alkoxy radicals, C₁-C₄ mesylaminoalkoxy radicals and carbamoylamino(C₁-C₄)-alkoxy radicals,
- R₄ is chosen from a hydrogen atom, halogen atoms, and C₁-C₄ alkyl radicals;

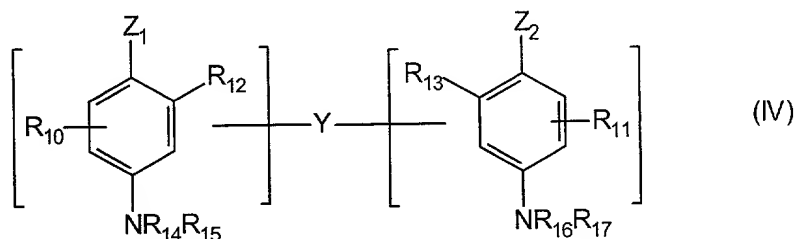
wherein at least one of the radicals R₁ to R₄ is other than a hydrogen atom.

53. The composition according to Claim 52, wherein said R₃ halogen atoms are chosen from chlorine, bromine, iodine and fluorine.

54. The composition according to Claim 52, wherein said para-phenylenediamine derivatives of formula (I) are chosen from para-toluylenediamine, 2-chloro-para-phenylenediamine, 2,3-dimethyl-para-phenylenediamine, 2,6-dimethyl-para-phenylenediamine, 2,6-diethyl-para-phenylenediamine, 2,5-dimethyl-para-phenylenediamine, N,N-dimethyl-para-phenylenediamine, N,N-diethyl-para-phenylenediamine, N,N-dipropyl-para-phenylenediamine, 4-amino-N,N-diethyl-3-methylaniline, N,N-bis(β-hydroxyethyl)-para-phenylenediamine, 4-amino-N,N-bis(β-hydroxyethyl)-2-methylaniline, 4-amino-2-chloro-N,N-bis(β-hydroxyethyl)aniline, 2-β-hydroxyethyl-para-phenylenediamine, 2-fluoro-para-phenylenediamine, 2-isopropyl-para-phenylenediamine, N-(β-hydroxypropyl)-para-phenylenediamine, 2-hydroxy-methyl-para-phenylenediamine, N,N-dimethyl-3-methyl-para-phenylenediamine,

N,N-(ethyl-β-hydroxyethyl)-para-phenylenediamine, N-(β,γ-dihydroxypropyl)-para-phenylenediamine, N-(4'-aminophenyl)-para-phenylenediamine, N-phenyl-para-phenylenediamine, 2-β-hydroxyethyloxy-para-phenylenediamine, 2-β-acetylaminomethoxy-para-phenylenediamine, N-(β-methoxyethyl)-para-phenylenediamine, and acid-addition salts thereof.

55. The composition according to Claim 32, wherein said double bases are chosen from compounds of formula (IV) below, and acid-addition salts thereof:



in which:

- Z_1 and Z_2 , which may be identical or different, are chosen from a hydroxyl radical and an $-NH_2$ radical which may be substituted with C_1 - C_4 alkyl radicals or with a linker arm Y;

- wherein said compounds of formula (IV) and salts thereof contain only one linker arm Y per molecule.

57. The composition according to Claim 55, wherein said double bases of formula (IV) are chosen from N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-aminophenyl)-1,3-diaminopropanol, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-aminophenyl)ethylenediamine, N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(4-methylaminophenyl)-tetramethylenediamine, N,N'-bis(ethyl)-N,N'-bis(4'-amino-3'-methylphenyl)ethylenedi-

amine, 1,8-bis(2,5-diaminophenoxy)-3,5-dioxaoctane, and acid-addition salts thereof.

58. The composition according to Claim 32, wherein said ortho-aminophenols are chosen from 2-aminophenol, 2-amino-5-methylphenol, 2-amino-6-methylphenol 5-acetamido-2-aminophenol, and acid-addition salts thereof.

59. The composition according to Claim 32, wherein said heterocyclic bases are chosen from pyridine compounds, pyrimidine compounds, pyrazole compounds, pyrazolopyrimidine compounds, and acid-addition salts thereof.

60. The composition according to Claim 32, wherein said at least one first oxidation base is present in an amount ranging from 0.0005 to 12% by weight relative to the total weight of the composition.

61. The composition according to Claim 60, wherein said at least one first oxidation base is present in an amount ranging from 0.005 to 6% by weight relative to the total weight of the composition.

62. The composition according to Claim 32, wherein said acid-addition salts are chosen from hydrochlorides, hydrobromides, sulphates, tartrates, lactates and acetates.

63. The composition according to Claim 32, wherein said composition further comprises water or a mixture of water and at least one organic solvent.

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64. The composition according to Claim 32, wherein said composition has a pH ranging from 5 to 11.

65. The composition according to Claim 32, further comprising at least one peroxidase.

66. A ready-to-use composition for the oxidation dyeing of keratin fibers, comprising:

- at least one first oxidation base chosen from

para-phenylenediamine compounds chosen from: para-toluylenediamine, 2-chloro-para-phenylenediamine, 2,3-dimethyl-para-phenylenediamine, 2,6-dimethyl-para-phenylenediamine, 2,6-diethyl-para-phenylenediamine, 2,5-dimethyl-para-phenylenediamine, N,N-dimethyl-para-phenylenediamine, N,N-diethyl-para-phenylenediamine, N,N-dipropyl-para-phenylenediamine, 4-amino-N,N-diethyl-3-methylaniline, N,N-bis(β -hydroxyethyl)-para-phenylenediamine, 4-amino-N,N-bis(β -hydroxyethyl)-2-methylaniline, 4-amino-2-chloro-N,N-bis(β -hydroxyethyl)aniline, 2- β -hydroxyethyl-para-phenylenediamine, 2-fluoro-para-phenylenediamine, 2-isopropyl-para-phenylenediamine, N-(β -hydroxypropyl)-para-phenylenediamine, 2-hydroxymethyl-para-phenylenediamine, N,N-dimethyl-3-methyl-para-phenylenediamine, N,N-(ethyl- β -hydroxyethyl)-para-phenylenediamine, N-(β , γ -dihydroxypropyl)-para-phenylenediamine, N-(4'-aminophenyl)-para-phenylenediamine, N-phenyl-para-phenylenediamine, 2- β -hydroxyethyloxy-para-phenylenediamine, 2- β -acetamino-

ethyloxy-para-phenylenediamine, N-(β -methoxyethyl)-para-phenylenediamine, and acid-addition salts thereof,

double bases chosen from: N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-aminophenyl)-1,3-diaminopropanol, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-aminophenyl)ethylenediamine, N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(4-methylaminophenyl)tetramethylenediamine, N,N'-bis(ethyl)-N,N'-bis(4'-amino-3'-methylphenyl)ethylenediamine, 1,8-bis(2,5-diaminophenoxy)-3,5-dioxaoctane, and acid-addition salts thereof,

ortho-aminophenols chosen from: 2-aminophenol, 2-amino-5-methylphenol, 2-amino-6-methylphenol, 5-acetamido-2-aminophenol, and acid-addition salts thereof,

pyridine compounds chosen from: 2,5-diaminopyridine, 2-(4-methoxyphenyl)amino-3-amino-pyridine, 2,3-diamino-6-methoxypyridine, 2-(γ -methoxy-ethyl)amino-3-amino-6-methoxypyridine, 3,4-diaminopyridine, and acid-addition salts thereof,

pyrimidine compounds chosen from: 2,4,5,6-tetraamino-pyrimidine, 4-hydroxy-2,5,6-triaminopyrimidine, 2-hydroxy-4,5,6-triamino-pyrimidine, 2,4-dihydroxy-5,6-diaminopyrimidine, 2,5,6-tri-amino-pyrimidine, and acid-addition salts thereof,

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pyrazolopyrimidine compounds chosen from:

- pyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,5-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- pyrazolo[1,5-a]pyrimidine-3,5-diamine;
- 2,7-dimethylpyrazolo[1,5-a]pyrimidine-3,5-diamine;

- 3-aminopyrazolo[1,5-a]pyrimidin-7-ol;
- 3-aminopyrazolo[1,5-a]pyrimidin-5-ol;
- 2-(3-aminopyrazolo[1,5-a]pyrimidin-7-ylamino)ethanol;
- 2-(7-aminopyrazolo[1,5-a]pyrimidin-3-ylamino)ethanol;
- 2-[(3-aminopyrazolo[1,5-a]pyrimidin-7-yl)-(2-hydroxy-ethyl)amino]ethanol;
- 2-[(7-aminopyrazolo[1,5-a]pyrimidin-3-yl)-(2-hydroxy-ethyl)amino]ethanol;
- 5,6-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,6-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,5,N7,N7-tetramethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;

and addition salts thereof and the tautomeric forms thereof, when a tautomeric equilibrium exists;

- at least one second oxidation base chosen from para-aminophenol, 4-amino-3-methylphenol, 4-amino-3-fluorophenol, 4-amino-3-hydroxymethylphenol, 4-amino-2-methylphenol, 4-amino-2-hydroxymethylphenol, 4-amino-2-methoxymethylphenol, 4-amino-2-aminomethylphenol, 4-amino-2-(β -hydroxyethylaminomethyl)phenol, 4-amino-2-fluorophenol, and acid-addition salts thereof;
- at least one meta-aminophenol coupler chosen from meta-aminophenol, 5-amino-2-methoxyphenol, 5-amino-2-(β -hydroxyethyloxy)phenol, 5-amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-4-methoxy-2-methylphenol, 5-amino-4-methoxy-2-methylphenol,

5-amino-4-chloro-2-methylphenol, 5-amino-2,4-dimethoxyphenol, 5-(γ -hydroxypropylamino)-2-methylphenol and acid-addition salts thereof;

- at least one 2-electron oxidoreductase chosen from pyranose oxidases, glucose oxidases, glycerol oxidases, lactate oxidases, pyruvate oxidases and uricases; and
- at least one donor for said 2-electron oxidoreductase chosen from D-glucose, L-sorbose, D-xylose, glycerol, dihydroxyacetone, lactic acid and salts thereof; pyruvic acid and salts thereof; and uric acid and salts thereof.

67. A process for dyeing keratin fibers, comprising applying at least one ready-to-use dye composition for the oxidation dyeing of keratin fibers to said fibers and developing for a period of time sufficient to achieve desired coloration, wherein said ready-to-use dye composition comprises:

- at least one first oxidation base chosen from para-phenylenediamine compounds other than para-phenylenediamine; double bases; ortho-aminophenols; heterocyclic bases; and acid-addition salts thereof,
- at least one second oxidation base chosen from para-aminophenols and acid-addition salts thereof,
- at least one coupler chosen from meta-aminophenols and acid-addition salts thereof,
- at least one enzyme chosen from 2-electron oxidoreductases, and

- at least one donor for said at least one enzyme.

68. A process for dyeing keratin fibers, comprising applying at least one ready-to-use dye composition for the oxidation dyeing of keratin fibers to said fibers and developing for a period sufficient to achieve the desired coloration, wherein said ready-to-use dye composition comprises:

- at least one first oxidation base chosen from

para-phenylenediamine compounds chosen from: para-toluylenediamine, 2-chloro-para-phenylenediamine, 2,3-dimethyl-para-phenylenediamine, 2,6-dimethyl-para-phenylenediamine, 2,6-diethyl-para-phenylenediamine, 2,5-dimethyl-para-phenylenediamine, N,N-dimethyl-para-phenylenediamine, N,N-diethyl-para-phenylenediamine, N,N-dipropyl-para-phenylenediamine, 4-amino-N,N-diethyl-3-methylaniline, N,N-bis(β -hydroxyethyl)-para-phenylenediamine, 4-amino-N,N-bis(β -hydroxyethyl)-2-methylaniline, 4-amino-2-chloro-N,N-bis(β -hydroxyethyl)aniline, 2- β -hydroxyethyl-para-phenylenediamine, 2-fluoro-para-phenylenediamine, 2-isopropyl-para-phenylenediamine, N-(β -hydroxypropyl)-para-phenylenediamine, 2-hydroxy-methyl-para-phenylenediamine, N,N-dimethyl-3-methyl-para-phenylenediamine, N,N-(ethyl- β -hydroxyethyl)-para-phenylenediamine, N-(β , γ -dihydroxypropyl)-para-phenylenediamine, N-(4'-aminophenyl)-para-phenylenediamine, N-phenyl-para-phenylenediamine, 2- β -hydroxyethyloxy-para-phenylenediamine, 2- β -acetamino-

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ethyloxy-para-phenylenediamine, N-(β -methoxyethyl)-para-phenylenediamine, and acid-addition salts thereof,

double bases chosen from: N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-aminophenyl)-1,3-diaminopropanol, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-aminophenyl)ethylenediamine, N,N'-bis(4'-aminophenyl)tetramethylenediamine, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-aminophenyl)tetramethylenediamine, N,N'-bis(4-methylaminophenyl)tetramethylenediamine, N,N'-bis(ethyl)-N,N'-bis(4'-amino-3'-methylphenyl)ethylenediamine, 1,8-bis(2,5-diaminophenoxy)-3,5-dioxaoctane, and acid-addition salts thereof,

ortho-aminophenols chosen from: 2-aminophenol, 2-amino-5-methylphenol, 2-amino-6-methylphenol, 5-acetamido-2-aminophenol, and acid-addition salts thereof,

pyridine compounds chosen from: 2,5-diaminopyridine, 2-(4-methoxyphenyl)amino-3-amino-pyridine, 2,3-diamino-6-methoxypyridine, 2-(γ -methoxy-ethyl)amino-3-amino-6-methoxypyridine, 3,4-diaminopyridine, and acid-addition salts thereof,

pyrimidine compounds chosen from: 2,4,5,6-tetraamino-pyrimidine, 4-hydroxy-2,5,6-triaminopyrimidine, 2-hydroxy-4,5,6-triamino-pyrimidine, 2,4-dihydroxy-5,6-diaminopyrimidine, 2,5,6-tri-amino-pyrimidine, and acid-addition salts thereof,

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pyrazolopyrimidine compounds chosen from:

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- and addition salts thereof and the tautomeric forms thereof, when a tautomeric equilibrium exists;

- 20-

5-amino-4-chloro-2-methylphenol, 5-amino-2,4-dimethoxyphenol 5-(γ-hydroxypropylamino)-2-methylphenol, and acid-addition salts thereof;

- at least one 2-electron oxidoreductase chosen from pyranose oxidases, glucose oxidases, glycerol oxidases, lactate oxidases, pyruvate oxidases and uricases; and
- at least one donor for said 2-electron oxidoreductase chosen from D-glucose, L-sorbose, D-xylose, glycerol, dihydroxyacetone, lactic acid and salts thereof; pyruvic acid and salts thereof; and uric acid and salts thereof.

69. A process for dyeing keratin fibers, comprising:

separately storing a first composition,
separately storing a second composition,
thereafter mixing said first composition with said second composition,
applying said mixture to said fibers and
developing for a period of time sufficient to achieve the desired coloration,
wherein said first composition comprises:

- at least one first oxidation base chosen from para-phenylenediamine compounds other than para-phenylenediamine, double bases, ortho-aminophenols, heterocyclic bases, and acid-addition salts thereof;
- at least one second oxidation base chosen from para-aminophenols and acid-addition salts thereof; and

wherein said second composition comprises at least one 2-electron oxidoreductase and at least one donor for said at least one 2-electron oxidoreductase.

developing for a period sufficient to achieve the desired coloration,

para-phenylenediamine compounds chosen from: para-toluylenediamine, 2-chloro-para-phenylenediamine, 2,3-dimethyl-para-phenylenediamine, 2,6-dimethyl-para-phenylenediamine, 2,6-diethyl-para-phenylenediamine, 2,5-dimethyl-para-phenylenediamine, N,N-dimethyl-para-phenylenediamine, N,N-diethyl-para-phenylenediamine, N,N-dipropyl-para-phenylenediamine, 4-amino-N,N-diethyl-3-methylaniline, N,N-bis(β -hydroxyethyl)-para-phenylenediamine, 4-amino-N,N-bis(β -hydroxyethyl)-2-methylaniline, 4-amino-2-chloro-N,N-bis(β -hydroxyethyl)aniline, 2- β -

hydroxyethyl-para-phenylenediamine, 2-fluoro-para-phenylenediamine, 2-isopropyl-para-phenylenediamine, N-(β -hydroxypropyl)-para-phenylenediamine, 2-hydroxymethyl-para-phenylenediamine, N,N-dimethyl-3-methyl-para-phenylenediamine, N,N-(ethyl- β -hydroxyethyl)-para-phenylenediamine, N-(β,γ -dihydroxypropyl)-para-phenylenediamine, N-(4'-aminophenyl)-para-phenylenediamine, N-phenyl-para-phenylenediamine, 2- β -hydroxyethyloxy-para-phenylenediamine, 2- β -acetaminoethyloxy-para-phenylenediamine, N-(β -methoxyethyl)-para-phenylenediamine, and acid-addition salts thereof,

double bases chosen from: N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-aminophenyl)-1,3-diaminopropanol, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-aminophenyl)ethylenediamine, N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(4-methylaminophenyl)tetramethylenediamine, N,N'-bis(ethyl)-N,N'-bis(4'-amino-3'-methylphenyl)ethylenediamine, 1,8-bis(2,5-diaminophenoxy)-3,5-dioxaoctane, and acid-addition salts thereof,

ortho-aminophenols chosen from: 2-aminophenol, 2-amino-5-methylphenol, 2-amino-6-methylphenol, 5-acetamido-2-aminophenol, and acid-addition salts thereof,

pyridine compounds chosen from: 2,5-diaminopyridine, 2-(4-methoxyphenyl)amino-3-amino-pyridine, 2,3-diamino-6-methoxypyridine, 2-(γ -

pyrimidine compounds chosen from: 2,4,5,6-tetraamino-pyrimidine, 4-hydroxy-2,5,6-triaminopyrimidine, 2-hydroxy-4,5,6-triamino-pyrimidine, 2,4-dihydroxy-5,6-diaminopyrimidine, 2,5,6-tri-amino-pyrimidine, and acid-addition salts thereof.

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3,5-diamino-4-(γ -hydroxyethyl)amino-1-methyl-pyrazole, and acid-addition salts thereof,

pyrazolopyrimidine compounds chosen from:

- pyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,5-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- pyrazolo[1,5-a]pyrimidine-3,5-diamine;
- 2,7-dimethylpyrazolo[1,5-a]pyrimidine-3,5-diamine;
- 3-aminopyrazolo[1,5-a]pyrimidin-7-ol;
- 3-aminopyrazolo[1,5-a]pyrimidin-5-ol;
- 2-(3-aminopyrazolo[1,5-a]pyrimidin-7-ylamino)ethanol;
- 2-(7-aminopyrazolo[1,5-a]pyrimidin-3-ylamino)ethanol;
- 2-[(3-aminopyrazolo[1,5-a]pyrimidin-7-yl)-(2-hydroxy-ethyl)amino]ethanol;
- 2-[(7-aminopyrazolo[1,5-a]pyrimidin-3-yl)-(2-hydroxy-ethyl)amino]ethanol;
- 5,6-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,6-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,5,N7,N7-tetramethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;

and addition salts thereof and the tautomeric forms thereof, when a tautomeric equilibrium exists;

at least one second oxidation base chosen from para-aminophenol, 4-amino-3-methylphenol, 4-amino-3-fluorophenol, 4-amino-3-hydroxymethylphenol, 4-amino-2-

methylphenol, 4-amino-2-hydroxymethylphenol, 4-amino-2-methoxymethylphenol, 4-amino-2-aminomethylphenol, 4-amino-2-(β -hydroxyethylaminomethyl)phenol 4-amino-2-fluorophenol, and acid-addition salts thereof; and

at least one meta-aminophenol coupler chosen from meta-aminophenol, 5-amino-2-methoxyphenol, 5-amino-2-(β -hydroxyethyloxy)phenol, 5-amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-4-methoxy-2-methylphenol, 5-amino-4-methoxy-2-methylphenol, 5-amino-4-chloro-2-methylphenol, 5-amino-2,4-dimethoxyphenol and 5-(γ -hydroxypropylamino)-2-methylphenol and acid-addition salts thereof;

- wherein said second composition comprises:

at least one 2-electron oxidoreductase enzyme chosen from pyranose oxidases, glucose oxidases, glycerol oxidases, lactate oxidases, pyruvate oxidases and uricases; and

- at least one donor for said enzyme chosen from D-glucose, L-sorbose, D-xylose, glycerol, dihydroxyacetone, lactic acid and salts thereof; pyruvic acid and salts thereof; and uric acid and salts thereof.

71. A multi-compartment dyeing kit, comprising at least two separate compartments wherein a first compartment contains a first composition and a second compartment contains a second composition

- wherein said first composition comprises:

at least one first oxidation base chosen from para-phenylenediamine compounds other than para-phenylenediamine, double bases, ortho-aminophenols, heterocyclic bases, and acid-addition salts thereof;

at least one second oxidation base chosen from para-aminophenols and acid-addition salts thereof, and

at least one coupler chosen from meta-aminophenols and acid-addition salts thereof; and

- wherein said second composition comprises at least one 2-electron oxidoreductase enzyme and at least one donor for said at least one enzyme.

72. A multi-compartment dyeing kit, comprising at least two separate compartments wherein a first compartment contains a first composition and a second compartment contains a second composition,

- wherein said first composition comprises:

at least one first oxidation base chosen from:

para-phenylenediamine compounds chosen from: para-toluylenediamine, 2-chloro-para-phenylenediamine, 2,3-dimethyl-para-phenylenediamine, 2,6-dimethyl-para-phenylenediamine, 2,6-diethyl-para-phenylenediamine, 2,5-dimethyl-para-phenylenediamine, N,N-dimethyl-para-phenylenediamine, N,N-diethyl-para-phenylenediamine, N,N-dipropyl-para-phenylenediamine, 4-amino-N,N-diethyl-3-methylaniline, N,N-bis(β -hydroxyethyl)-para-phenylenediamine, 4-amino-N,N-bis(β -

hydroxyethyl)-2-methylaniline, 4-amino-2-chloro-N,N-bis(β -hydroxyethyl)aniline, 2- β -hydroxyethyl-para-phenylenediamine, 2-fluoro-para-phenylenediamine, 2-isopropyl-para-phenylenediamine, N-(β -hydroxypropyl)-para-phenylenediamine, 2-hydroxy-methyl-para-phenylenediamine, N,N-dimethyl-3-methyl-para-phenylenediamine, N,N-(ethyl- β -hydroxyethyl)-para-phenylenediamine, N-(β,γ -dihydroxypropyl)-para-phenylenediamine, N-(4'-aminophenyl)-para-phenylenediamine, N-phenyl-para-phenylenediamine, 2- β -hydroxyethyloxy-para-phenylenediamine, 2- β -acetylamino-ethyloxy-para-phenylenediamine, N-(β -methoxyethyl)-para-phenylenediamine, and acid-addition salts thereof,

double bases chosen from: N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-amino-phenyl)-1,3-diaminopropanol, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-amino-phenyl)ethylenediamine, N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(4-methylaminophenyl)tetramethylenediamine, N,N'-bis(ethyl)-N,N'-bis(4'-amino-3'-methylphenyl)ethylenediamine, 1,8-bis(2,5-diaminophenoxy)-3,5-dioxaoctane, and acid-addition salts thereof,

ortho-aminophenols chosen from: 2-aminophenol, 2-amino-5-methylphenol, 2-amino-6-methylphenol, 5-acetamido-2-aminophenol, and acid-addition salts thereof,

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pyridine compounds chosen from: 2,5-diaminopyridine,
2-(4-methoxyphenyl)amino-3-amino-pyridine, 2,3-diamino-6-methoxypyridine, 2-(γ -methoxy-ethyl)amino-3-amino-6-methoxypyridine, 3,4-diaminopyridine, and acid-addition salts thereof,

pyrimidine compounds chosen from: 2,4,5,6-tetraamino-pyrimidine,
4-hydroxy-2,5,6-triaminopyrimidine, 2-hydroxy-4,5,6-triamino-pyrimidine,
2,4-dihydroxy-5,6-diaminopyrimidine, 2,5,6-tri-amino-pyrimidine, and acid-addition salts thereof,

pyrazole compounds chosen from: 4,5-diamino-1-methyl-pyrazole,
3,4-diaminopyrazole, 4,5-diamino-1-(4'-chlorobenzyl)pyrazole,
4,5-diamino-1,3-dimethyl-pyrazole, 4,5-diamino-3-methyl-1-phenylpyrazole, 4,5-diamino-1-methyl-3-phenylpyrazole, 4-amino-1,3-di-methyl-5-hydrazinopyrazole,
1-benzyl-4,5-diamino-3-methyl-pyrazole, 4,5-diamino-3-tert-butyl-1-methyl-pyrazole,
4,5-diamino-1-tert-butyl-3-methylpyrazole,
4,5-diamino-1-(γ -hydroxyethyl)-3-methylpyrazole, 4,5-diamino-1-ethyl-3-methylpyrazole, 4,5-diamino-1-ethyl-3-(4'-methoxyphenyl)pyrazole, 4,5-diamino-1-ethyl-3-hydroxymethylpyrazole,
4,5-diamino-3-hydroxymethyl-1-methyl-pyrazole, 4,5-diamino-3-hydroxymethyl-1-iso-propyl-pyrazole, 4,5-diamino-3-methyl-1-isopropyl-pyrazole,
4-amino-5-(2'-amino-ethyl)amino-1,3-dimethyl-pyrazole, 3,4,5-triamino-pyrazole,

1-methyl-3,4,5-tri-amino-pyrazole, 3,5-diamino-1-methyl-4-methylamino-pyrazole, 3,5-diamino-4-(γ -hydroxyethyl)amino-1-methyl-pyrazole, and acid-addition salts thereof,

pyrazolopyrimidine compounds chosen from:

- pyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,5-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- pyrazolo[1,5-a]pyrimidine-3,5-diamine;
- 2,7-dimethylpyrazolo[1,5-a]pyrimidine-3,5-diamine;
- 3-aminopyrazolo[1,5-a]pyrimidin-7-ol;
- 3-aminopyrazolo[1,5-a]pyrimidin-5-ol;
- 2-(3-aminopyrazolo[1,5-a]pyrimidin-7-ylamino)ethanol;
- 2-(7-aminopyrazolo[1,5-a]pyrimidin-3-ylamino)ethanol;
- 2-[(3-aminopyrazolo[1,5-a]pyrimidin-7-yl)-(2-hydroxy-ethyl)amino]ethanol;
- 2-[(7-aminopyrazolo[1,5-a]pyrimidin-3-yl)-(2-hydroxy-ethyl)amino]ethanol;
- 5,6-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,6-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,5,N7,N7-tetramethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;

and addition salts thereof and the tautomeric forms thereof, when a tautomeric equilibrium exists;

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at least one meta-aminophenol coupler chosen from meta-aminophenol, 5-amino-2-methoxyphenol, 5-amino-2-(β -hydroxyethyloxy)phenol, 5-amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-4-methoxy-2-methylphenol, 5-amino-4-methoxy-2-methylphenol, 5-amino-4-chloro-2-methylphenol, 5-amino-2,4-dimethoxyphenol and 5-(γ -hydroxypropylamino)-2-methylphenol and acid-addition salts thereof;

at least one 2-electron oxidoreductase enzyme chosen from pyranose oxidases, glucose oxidases, glycerol oxidases, lactate oxidases, pyruvate oxidases and uricases; and

73. A ready-to-use composition for the oxidation dyeing of keratin fibers, comprising,

- 2-β-hydroxyethyl-para-phenylenediamine di-hydrochloride,
- para-aminophenol,
- at least one coupler chosen from meta-aminophenol and 2-methyl-5-aminophenol,
- uricase,
- uric acid.

74. The composition according to Claim 32, further comprising at least one direct dye. --

REMARKS

Claims 1 to 31 have been canceled and replaced by new claims 32 to 74. Support for these claims can be found in the original specification and claims. No new matter has been added. Applicants now await an action on the merits.

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-33-

COMPOSITION FOR THE OXIDATION DYEING OF KERATIN
FIBRES AND DYEING PROCESS USING THIS COMPOSITION

The invention relates to a composition for the oxidation dyeing of keratin fibres, and in particular human keratin fibres such as the hair, comprising, in a medium which is suitable for dyeing, at least one first oxidation base chosen from para-phenylenediamine derivatives, double bases, ortho-aminophenols and heterocyclic bases, at least one second oxidation base chosen from para-aminophenols, at least one meta-aminophenol as coupler and at least one enzyme of 2-electron oxidoreductase type in the presence of at least one donor for the said enzyme, and to the dyeing process using this composition.

It is known to dye keratin fibres, and in particular human hair, with dye compositions containing oxidation dye precursors, in particular ortho- or para-phenylenediamines, ortho- or para-aminophenols and heterocyclic bases, which are generally referred to as oxidation bases. Oxidation dye precursors, or oxidation bases, are colourless or weakly coloured compounds which, when combined with oxidizing products, can give rise to coloured compounds and dyes by a process of oxidative condensation.

It is also known that the shades obtained with these oxidation bases can be varied by combining them with couplers or colour modifiers, the latter being chosen in particular from aromatic meta-diamines,

meta-aminophenols, meta-diphenols and certain heterocyclic compounds.

The variety of molecules used as oxidation bases and couplers allows a wide range of colours to be
5 obtained.

The so-called "permanent" coloration obtained by means of these oxidation dyes must moreover satisfy a certain number of requirements. Thus, it must have no toxicological drawbacks, it must be able to give shades
10 of the desired intensity and it must be able to withstand external agents (light, bad weather, washing, permanent-waving, perspiration, rubbing).

The dyes must also be able to cover white hair and, lastly, they must be as unselective as
15 possible, i.e. they must give the smallest possible colour differences along the same length of keratin fibre, which may in fact be differently sensitized (i.e. damaged) between its tip and its root.

The oxidation dyeing of keratin fibres is
20 generally carried out in alkaline medium, in the presence of hydrogen peroxide. However, the use of alkaline media in the presence of hydrogen peroxide have the drawback of causing appreciable degradation of the fibres, as well as considerable bleaching of the
25 keratin fibres, which is not always desirable.

The oxidation dyeing of keratin fibres can also be carried out using oxidizing systems other than hydrogen peroxide, such as enzymatic systems. Thus, it

has already been proposed to dye keratin fibres, in particular in patent application EP-A-0,310,675, with compositions comprising an oxidation base and optionally a coupler, in combination with enzymes such as pyranose oxidase, glucose oxidase or uricase, in the presence of a donor for the said enzymes. Although being used under conditions which do not result in a degradation of the keratin fibres which is comparable to that caused by the dyes used in the presence of hydrogen peroxide, these dyeing processes nevertheless lead to colorations which are not entirely satisfactory, in particular as regards their intensity and resistance to the various attacking factors to which the hair may be subjected.

The Applicant has now discovered that it is possible to obtain new dyes, which are capable of leading to intense and chromatic colorations, without giving rise to any significant degradation of the keratin fibres, and which are relatively unselective and show good resistance to the various attacking factors to which the hair may be subjected, by combining at least one first oxidation base chosen from para-phenylenediamine derivatives other than para-phenylenediamine, double bases, ortho-aminophenols and heterocyclic bases, at least one second oxidation base chosen from para-aminophenols, at least one meta-aminophenol as coupler and at least one enzyme of 2-electron oxidoreductase type in the presence of at

least one donor for the said enzyme.

This discovery forms the basis of the present invention.

A first subject of the invention is thus a
5 ready-to-use composition for the oxidation dyeing of
keratin fibres, and in particular human keratin fibres
such as the hair, characterized in that it comprises,
in a medium which is suitable for dyeing:
- at least one first oxidation base chosen from para-
10 phenylenediamine derivatives other than para-
phenylenediamine, double bases, ortho-aminophenols and
heterocyclic bases,
- at least one second oxidation base chosen from para-
aminophenols,
15 - at least one meta-aminophenol as coupler,
- at least one enzyme of 2-electron oxidoreductase
type, and
- at least one donor for the said enzyme.

The ready-to-use dye composition in
20 accordance with the invention leads to intense
relatively unselective colorations with excellent
properties of resistance both to atmospheric agents
such as light and bad weather and to perspiration and
the various treatments to which the hair may be
25 subjected (washing, permanent-waving).

A subject of the invention is also a process
for the oxidation dyeing of keratin fibres using this
ready-to-use dye composition.

The 2-electron oxidoreductase(s) used in the ready-to-use dye composition in accordance with the invention can be chosen in particular from pyranose oxidases, glucose oxidases, glycerol oxidases, lactate
5 oxidases, pyruvate oxidases and uricases.

According to the invention, the 2-electron oxidoreductase is preferably chosen from uricases of animal, microbiological or biotechnological origin.

By way of example, mention may be made of
10 uricase extracted from boar liver, uricase from *Arthrobacter globiformis*, as well as uricase from *Aspergillus flavus*.

The 2-electron oxidoreductase(s) can be used in pure crystalline form or in a form diluted in a
15 diluent which is inert with respect to the said 2-electron oxidoreductase.

The 2-electron oxidoreductase(s) in accordance with the invention preferably represent(s) from 0.01 to 20% by weight approximately relative to
20 the total weight of the ready-to-use dye composition, and even more preferably from 0.1 to 5% by weight approximately relative to this weight.

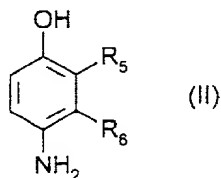
According to the invention, the term donor is understood to refer to the various substrates involved
25 in the functioning of the said 2-electron oxidoreductase(s).

The nature of the donor (or substrate) for the said enzyme varies depending on the nature of the

2-electron oxidoreductase used. For example, as donors for the pyranose oxidases, mention may be made of D-glucose, L-sorbose and D-xylose; as a donor for the glucose oxidases, mention may be made of D-glucose; as
 5 donors for the glycerol oxidases, mention may be made of glycerol and dihydroxyacetone; as donors for the lactate oxidases, mention may be made of lactic acid and its salts; as donors for the pyruvate oxidases, mention may be made of pyruvic acid and its salts; and
 10 lastly, as donors for the uricases, mention may be made of uric acid and its salts.

The donor(s) (or substrate(s)) used in accordance with the invention preferably represent(s) from 0.01 to 20% by weight approximately relative to
 15 the total weight of the ready-to-use dye composition in accordance with the invention, and even more preferably from 0.1 to 5% by approximately relative to this weight.

Among the para-aminophenols which can be used
 20 as second oxidation base in the dye compositions according to the invention, mention may be made in particular of the compounds corresponding to formula (II) below, and the addition salts thereof with an acid:



in which:

- R_5 represents a hydrogen or halogen atom or a C_1 - C_4 alkyl, C_1 - C_4 monohydroxyalkyl, $(C_1$ - C_4)alkoxy(C_1 - C_4)alkyl, C_1 - C_4 aminoalkyl or hydroxy(C_1 - C_4)alkylamino(C_1 - C_4)alkyl radical,
 - 5 - R_6 represents a hydrogen or halogen atom or a C_1 - C_4 alkyl, C_1 - C_4 monohydroxyalkyl, C_2 - C_4 polyhydroxyalkyl, C_1 - C_4 aminoalkyl, cyano(C_1 - C_4)alkyl or $(C_1$ - C_4)alkoxy(C_1 - C_4)alkyl radical,
- it being understood that at least one of the radicals R_5
- 10 or R_6 represents a hydrogen atom.

Among the para-aminophenols of formula (II) above, mention may be made more particularly of para-aminophenol, 4-amino-3-methylphenol, 4-amino-3-fluorophenol, 4-amino-3-hydroxymethylphenol, 4-amino-2-methylphenol, 4-amino-2-hydroxymethylphenol, 4-amino-2-methoxymethylphenol, 4-amino-2-aminomethylphenol, 4-amino-2-(β -hydroxyethylaminomethyl)phenol and 4-amino-2-fluorophenol, and the addition salts thereof with an acid.

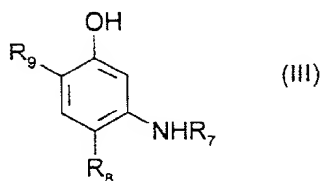
15

20 The para-aminophenol(s) which can be used as second oxidation base preferably represent(s) from 0.0005 to 12% by weight approximately relative to the total weight of the ready-to-use dye composition in accordance with the invention, and even more preferably

25 from 0.005 to 6% by weight approximately relative to this weight.

The meta-aminophenol(s) which can be used as coupler in the ready-to-use dye composition in accordance with the invention is (are) preferably chosen

from the compounds of formula (III) below, and the addition salts thereof with an acid:



in which:

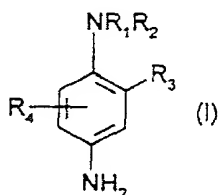
- R_7 represents a hydrogen atom or a C_1 - C_4 alkyl, C_1 - C_4 monohydroxyalkyl or C_2 - C_4 polyhydroxyalkyl radical,
- R_8 represents a hydrogen atom, a C_1 - C_4 alkyl or C_1 - C_4 alkoxy radical or a halogen atom chosen from chlorine, bromine and fluorine,
- R_9 represents a hydrogen atom or a C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 monohydroxyalkyl, C_2 - C_4 polyhydroxyalkyl, C_1 - C_4 monohydroxyalkoxy or C_2 - C_4 polyhydroxyalkoxy radical.

Among the meta-aminophenols of formula (III) above, mention may be made more particularly of meta-aminophenol, 5-amino-2-methoxyphenol, 5-amino-2-(β -hydroxyethyloxy)phenol, 5-amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-4-methoxy-2-methylphenol, 5-amino-4-methoxy-2-methylphenol, 5-amino-4-chloro-2-methylphenol, 5-amino-2,4-dimethoxyphenol and 5-(γ -hydroxypropylamino)-2-methylphenol, and the addition salts thereof with an acid.

The meta-aminophenol(s) which can be used as coupler preferably represent(s) from 0.0001 to 8% by weight approximately relative to the total weight of the

ready-to-use dye composition, and even more preferably from 0.005 to 5% by weight approximately relative to this weight.

Among the para-phenylenediamine derivatives which can be used as first oxidation base in the ready-to-use dye composition in accordance with the invention, mention may be made in particular of the compounds of formula (I) below, and the addition salts thereof with an acid:



in which:

- R_1 represents a hydrogen atom, a $\text{C}_1\text{-C}_4$ alkyl radical, a $\text{C}_1\text{-C}_4$ monohydroxyalkyl radical, a $\text{C}_2\text{-C}_4$ polyhydroxyalkyl radical, a $(\text{C}_1\text{-C}_4)$ alkoxy $(\text{C}_1\text{-C}_4)$ alkyl radical, a $\text{C}_1\text{-C}_4$ alkyl radical substituted with a nitrogenous group, a phenyl radical or a 4'-aminophenyl radical;
- R_2 represents a hydrogen atom, a $\text{C}_1\text{-C}_4$ alkyl radical, a $\text{C}_1\text{-C}_4$ monohydroxyalkyl radical, a $\text{C}_2\text{-C}_4$ polyhydroxyalkyl radical, a $(\text{C}_1\text{-C}_4)$ alkoxy $(\text{C}_1\text{-C}_4)$ alkyl radical or a $\text{C}_1\text{-C}_4$ alkyl radical substituted with a nitrogenous group;
- R_3 represents a hydrogen atom, a halogen atom such as a chlorine, bromine, iodine or fluorine atom, a $\text{C}_1\text{-C}_4$ alkyl radical, a $\text{C}_1\text{-C}_4$ monohydroxyalkyl radical, a $\text{C}_1\text{-C}_4$ hydroxyalkoxy radical, an acetylamino $(\text{C}_1\text{-C}_4)$ -

alkoxy radical, a C₁-C₄ mesylaminoalkoxy radical or a carbamoylamino(C₁-C₄)alkoxy radical,

- R₄ represents a hydrogen or halogen atom or a C₁-C₄ alkyl radical;

5 it being understood that at least one of the radicals R₁ to R₄ is other than a hydrogen atom.

Among the nitrogenous groups of formula (I) above, mention may be made in particular of amino, mono(C₁-C₄)alkylamino, di(C₁-C₄)alkylamino, tri(C₁-C₄)alkylamino, monohydroxy(C₁-C₄)alkylamino, imidazolinium and ammonium radicals.

Among the para-phenylenediamine derivatives of formula (I) above, mention may be made more particularly of para-toluylenediamine, 2-chloro-para-phenylenediamine, 2,3-dimethyl-para-phenylenediamine, 2,6-dimethyl-para-phenylenediamine, 2,6-diethyl-para-phenylenediamine, 2,5-dimethyl-para-phenylenediamine, N,N-dimethyl-para-phenylenediamine, N,N-diethyl-para-phenylenediamine, N,N-dipropyl-para-phenylenediamine, 4-amino-N,N-diethyl-3-methylaniline, N,N-bis(β-hydroxyethyl)-para-phenylenediamine, 4-amino-N,N-bis(β-hydroxyethyl)-2-methylaniline, 4-amino-2-chloro-N,N-bis(β-hydroxyethyl)aniline, 2-β-hydroxyethyl-para-phenylenediamine, 2-fluoro-para-phenylenediamine, 2-isopropyl-para-phenylenediamine, N-(β-hydroxypropyl)-para-phenylenediamine, 2-hydroxymethyl-para-phenylenediamine, N,N-dimethyl-3-methyl-para-phenylenediamine, N,N-(ethyl-β-hydroxyethyl)-para-phenylenediamine, N-(β,γ-dihydroxypropyl)-para-phenylene-

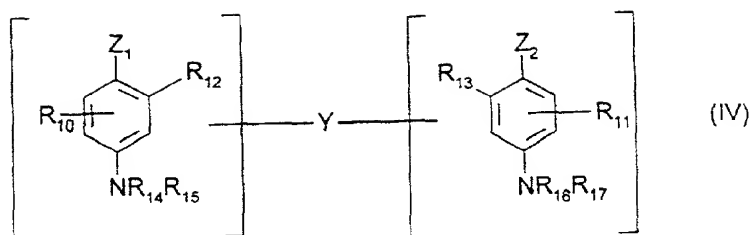
diamine, N-(4'-aminophenyl)-para-phenylenediamine, N-phenyl-para-phenylenediamine, 2- β -hydroxyethyloxy-para-phenylenediamine, 2- β -acetylaminoethyloxy-para-phenylenediamine and N-(β -methoxyethyl)-para-

5 phenylenediamine, and the addition salts thereof with an acid.

Among the para-phenylenediamine derivatives of formula (I) above, para-toluylenediamine, 2-isopropyl-para-phenylenediamine, 2- β -hydroxyethyl-para-phenylene-
10 diamine, 2- β -hydroxyethyloxy-para-phenylenediamine, 2,6-dimethyl-para-phenylenediamine, 2,6-diethyl-para-phenylenediamine, 2,3-dimethyl-para-phenylenediamine, N,N-bis(β -hydroxyethyl)-para-phenylenediamine, 2-chloro-para-phenylenediamine and 2- β -acetylaminoethyloxy-para-
15 phenylenediamine and the addition salts thereof with an acid are most particularly preferred.

According to the invention, the term double bases is understood to refer to the compounds containing at least two aromatic rings bearing amino
20 and/or hydroxyl groups.

Among the double bases which can be used as first oxidation base in the ready-to-use dye composition in accordance with the invention, mention may be made in particular of the compounds of formula
25 (IV) below, and the addition salts thereof with an acid:



in which:

- Z_1 and Z_2 , which may be identical or different, represent a hydroxyl or -NH_2 radical which may be substituted with a $\text{C}_1\text{-C}_4$ alkyl radical or with a linker arm Y;
 - the linker arm Y represents a linear or branched alkylene chain containing from 1 to 14 carbon atoms, which may be interrupted by or terminated with one or more nitrogenous groups and/or one or more hetero atoms such as oxygen, sulphur or nitrogen atoms, and optionally substituted with one or more hydroxyl or $\text{C}_1\text{-C}_6$ alkoxy radicals;
 - R_{10} and R_{11} represent a hydrogen or halogen atom, a $\text{C}_1\text{-C}_4$ alkyl radical, a $\text{C}_1\text{-C}_4$ monohydroxyalkyl radical, a $\text{C}_2\text{-C}_4$ polyhydroxyalkyl radical, a $\text{C}_1\text{-C}_4$ aminoalkyl radical or a linker arm Y;
 - R_{12} , R_{13} , R_{14} , R_{15} , R_{16} and R_{17} , which may be identical or different, represent a hydrogen atom, a linker arm Y or a $\text{C}_1\text{-C}_4$ alkyl radical;
- it being understood that the compounds of formula (IV) contain only one linker arm Y per molecule.

Among the nitrogenous groups of formula (IV) above, mention may be made in particular of amino,

mono(C₁-C₄)alkylamino, di(C₁-C₄)alkylamino, tri(C₁-C₄)alkylamino, monohydroxy(C₁-C₄)alkylamino, imidazolinium and ammonium radicals.

Among the double bases of formula (IV) above, mention may be made more particularly of N,N'-bis-(β-hydroxyethyl)-N,N'-bis(4'-aminophenyl)-1,3-diaminopropanol, N,N'-bis(β-hydroxyethyl)-N,N'-bis(4'-aminophenyl)ethylenediamine, N,N'-bis(4-aminophenyl)-tetramethylenediamine, N,N'-bis(β-hydroxyethyl)-N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(4-methylaminophenyl)tetramethylenediamine, N,N'-bis(ethyl)-N,N'-bis(4'-amino-3'-methylphenyl)-ethylenediamine and 1,8-bis(2,5-diaminophenoxy)-3,5-dioxaoctane, and the addition salts thereof with an acid.

Among these double bases of formula (IV), N,N'-bis(β-hydroxyethyl)-N,N'-bis(4'-aminophenyl)-1,3-diaminopropanol and 1,8-bis(2,5-diaminophenoxy)-3,5-dioxaoctane, or one of the addition salts thereof with an acid, are particularly preferred.

Among the ortho-aminophenols which can be used as first oxidation base in the ready-to-use dye composition in accordance with the invention, mention may be made more particularly of 2-aminophenol, 2-amino-5-methylphenol, 2-amino-6-methylphenol and 5-acetamido-2-aminophenol, and the addition salts thereof with an acid.

Among the heterocyclic bases which can be

used as first oxidation base in the dye composition in accordance with the invention, mention may be made more particularly of pyridine derivatives, pyrimidine derivatives, pyrazole derivatives and pyrazolo-
5 pyrimidine derivatives, and the addition salts thereof with an acid.

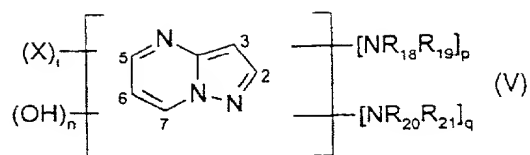
Among the pyridine derivatives, mention may be made more particularly of the compounds described, for example, in patents GB 1,026,978 and GB 1,153,196,
10 such as 2,5-diaminopyridine, 2-(4-methoxyphenyl)amino-3-aminopyridine, 2,3-diamino-6-methoxypyridine, 2-(β -methoxyethyl)amino-3-amino-6-methoxypyridine and 3,4-diaminopyridine, and the addition salts thereof with an acid.

15 Among the pyrimidine derivatives, mention may be made more particularly of the compounds described, for example, in German patent DE 2,359,399 or Japanese patents JP 88-169,571 and JP 91-10659 or patent applications WO 96/15765, such as 2,4,5,6-tetraamino-
20 pyrimidine, 4-hydroxy-2,5,6-triaminopyrimidine, 2-hydroxy-4,5,6-triaminopyrimidine, 2,4-dihydroxy-5,6-diaminopyrimidine and 2,5,6-triaminopyrimidine, and the addition salts thereof with an acid.

Among the pyrazole derivatives, mention may
25 be made more particularly of the compounds described in patents DE 3,843,892, DE 4,133,957 and patent applications WO 94/08969, WO 94/08970, FR-A-2,733,749 and DE 195 43 988, such as 4,5-diamino-1-methyl-

pyrazole, 3,4-diaminopyrazole, 4,5-diamino-
 1-(4'-chlorobenzyl)pyrazole, 4,5-diamino-1,3-dimethyl-
 pyrazole, 4,5-diamino-3-methyl-1-phenylpyrazole,
 4,5-diamino-1-methyl-3-phenylpyrazole, 4-amino-1,3-di-
 5 methyl-5-hydrazinopyrazole, 1-benzyl-4,5-diamino-
 3-methylpyrazole, 4,5-diamino-3-tert-butyl-1-methyl-
 pyrazole, 4,5-diamino-1-tert-butyl-3-methylpyrazole,
 4,5-diamino-1-(β -hydroxyethyl)-3-methylpyrazole,
 4,5-diamino-1-ethyl-3-methylpyrazole, 4,5-diamino-
 10 1-ethyl-3-(4'-methoxyphenyl)pyrazole, 4,5-diamino-
 1-ethyl-3-hydroxymethylpyrazole, 4,5-diamino-
 3-hydroxymethyl-1-methylpyrazole, 4,5-diamino-
 3-hydroxymethyl-1-isopropylpyrazole, 4,5-diamino-
 3-methyl-1-isopropylpyrazole, 4-amino-5-(2'-amino-
 15 ethyl)amino-1,3-dimethylpyrazole, 3,4,5-triamino-
 pyrazole, 1-methyl-3,4,5-triaminopyrazole, 3,5-diamino-
 1-methyl-4-methylaminopyrazole and 3,5-diamino-
 4-(β -hydroxyethyl)amino-1-methylpyrazole, and the
 addition salts thereof with an acid.

20 Among the pyrazolopyrimidine derivatives,
 mention may be made more particularly of the
 pyrazolo[1,5-a]pyrimidines of formula (V) below, and
 the addition salts thereof with an acid or with a base
 and the tautomeric forms thereof, when a tautomeric
 25 equilibrium exists:



in which:

- R_{18} , R_{19} , R_{20} and R_{21} , which may be identical or different, denote a hydrogen atom, a C_1 - C_4 alkyl radical, an aryl radical, a C_1 - C_4 hydroxyalkyl radical, a C_2 - C_4 polyhydroxyalkyl radical, a $(C_1$ - $C_4)$ alkoxy(C_1 - C_4)-alkyl radical, a C_1 - C_4 aminoalkyl radical (it being possible for the amine to be protected with an acetyl, ureido or sulphonyl radical), a $(C_1$ - $C_4)$ alkylamino(C_1 - C_4)-alkyl radical, a di[(C_1 - C_4)alkyl]amino(C_1 - C_4)alkyl radical (it being possible for the dialkyl radicals to form a 5- or 6-membered carbon-based ring or a heterocycle), a hydroxy(C_1 - C_4)alkyl- or di[hydroxy-(C_1 - C_4)alkyl]amino(C_1 - C_4)alkyl radical;
- the radicals X, which may be identical or different, denote a hydrogen atom, a C_1 - C_4 alkyl radical, an aryl radical, a C_1 - C_4 hydroxyalkyl radical, a C_2 - C_4 polyhydroxyalkyl radical, a C_1 - C_4 aminoalkyl radical, a $(C_1$ - $C_4)$ alkylamino(C_1 - C_4)alkyl radical, a di[(C_1 - C_4)-alkyl]amino(C_1 - C_4)alkyl radical (it being possible for the dialkyls to form a 5- or 6-membered carbon-based ring or a heterocycle), a hydroxy(C_1 - C_4)alkyl- or di-[hydroxy(C_1 - C_4)alkyl]amino(C_1 - C_4)alkyl radical, an amino radical, a $(C_1$ - C_4)alkyl- or di[(C_1 - C_4)alkyl]amino radical; a halogen atom, a carboxylic acid group, a

sulphonic acid group;

- i is equal to 0, 1, 2 or 3;

- p is equal to 0 or 1;

- q is equal to 0 or 1;

5 - n is equal to 0 or 1;

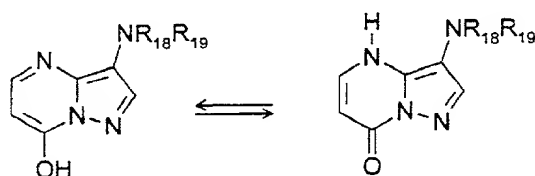
with the proviso that:

- the sum $p + q$ is other than 0;

- when $p + q$ is equal to 2, then n is equal to 0 and
the groups $NR_{18}R_{19}$ and $NR_{20}R_{21}$ occupy the (2,3); (5,6);
10 (6,7); (3,5) or (3,7) positions;

- when $p + q$ is equal to 1, then n is equal to 1 and
the group $NR_{18}R_{19}$ (or $NR_{20}R_{21}$) and the OH group occupy
the (2,3); (5,6); (6,7); (3,5) or (3,7) positions.

When the pyrazolo[1,5-a]pyrimidines of formula
15 (V) above are such that they contain a hydroxyl group
on one of the positions 2, 5 or 7 α to a nitrogen atom,
a tautomeric equilibrium exists represented, for
example, by the following scheme:



Among the pyrazolo[1,5-a]pyrimidines of formula
20 (V) above, mention may be made in particular of:

- pyrazolo[1,5-a]pyrimidine-3,7-diamine;

- 2,5-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;

- pyrazolo[1,5-a]pyrimidine-3,5-diamine;

- 2,7-dimethylpyrazolo[1,5-a]pyrimidine-3,5-diamine;

- 3-aminopyrazolo[1,5-a]pyrimidin-7-ol;
- 3-aminopyrazolo[1,5-a]pyrimidin-5-ol;
- 2-(3-aminopyrazolo[1,5-a]pyrimidin-7-ylamino)ethanol;
- 2-(7-aminopyrazolo[1,5-a]pyrimidin-3-ylamino)ethanol;
- 5 - 2-[(3-aminopyrazolo[1,5-a]pyrimidin-7-yl)-(2-hydroxy-ethyl)amino]ethanol;
- 2-[(7-aminopyrazolo[1,5-a]pyrimidin-3-yl)-(2-hydroxy-ethyl)amino]ethanol;
- 5,6-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 10 - 2,6-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,5,N7,N7-tetramethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;

and the addition salts thereof and the tautomeric forms thereof, when a tautomeric equilibrium exists.

- 15 The pyrazolo[1,5-a]pyrimidines of formula (V) above can be prepared by cyclization starting with an aminopyrazole, according to the syntheses described in the following references:

- EP 628559 Beiersdorf-Lilly.
- 20 - R. Vishdu, H. Navedul, Indian J. Chem., 34b (6), 514, 1995.
- N.S. Ibrahim, K.U. Sadek, F.A. Abdel-Al, Arch. Pharm., 320, 240, 1987.
- R.H. Springer, M.B. Scholten, D.E. O'Brien,
- 25 T. Novinson, J.P. Miller, R.K. Robins, J. Med. Chem., 25, 235, 1982.
- T. Novinson, R.K. Robins, T.R. Matthews, J. Med. Chem., 20, 296, 1977.

- US 3907799 ICN Pharmaceuticals.

The pyrazolo[1,5-a]pyrimidines of formula (V) above can also be prepared by cyclization starting from hydrazine, according to the syntheses described in the following references:

- A. McKillop and R.J. Kobilecki, Heterocycles, 6(9), 1355, 1977.
- E. Alcade, J. De Mendoza, J.M. Marcia-Marquina, C. Almera, J. Elguero, J. Heterocyclic Chem., 11(3), 423, 1974.
- K. Saito, I. Hori, M. Higarashi, H. Midorikawa, Bull. Chem. Soc. Japan, 47(2), 476, 1974.

The para-phenylenediamine derivative(s) and/or the double base(s) and/or the ortho-aminophenol(s) and/or the heterocyclic base(s) which can be used as first oxidation base preferably represent(s) from 0.0005 to 12% by weight approximately relative to the total weight of the ready-to-use dye composition according to the invention, and even more preferably from 0.005 to 6% by weight approximately relative to this weight.

The ready-to-use dye composition in accordance with the invention can also contain one or more additional couplers other than meta-aminophenols used according to the invention and/or one or more direct dyes, in particular in order to modify the shades or to enrich them with glints.

Among the couplers which can be present

additionally in the ready-to-use dye composition in accordance with the invention, mention may be made in particular of meta-phenylenediamines, meta-diphenols and heterocyclic couplers, and the addition salts thereof with an acid.

When they are present, these additional couplers preferably represent from 0.0001 to 10% by weight approximately relative to the total weight of the ready-to-use dye composition, and even more preferably from 0.005 to 5% by weight approximately relative to this weight.

In general, the addition salts with an acid which can be used in the context of the dye compositions of the invention (oxidation bases and couplers) are chosen in particular from the hydrochlorides, hydrobromides, sulphates, tartrates, lactates and acetates.

The medium which is suitable for dyeing (or support) for the ready-to-use dye composition in accordance with the invention generally consists of water or a mixture of water and at least one organic solvent to dissolve the compounds which would not be sufficiently soluble in water. By way of organic solvents, mention may be made, for example, of C₁-C₄ alkanols, such as ethanol and isopropanol; glycerol; glycols and glycol ethers such as 2-butoxyethanol, propylene glycol, propylene glycol monomethyl ether, diethylene glycol monoethyl ether and monomethyl ether,

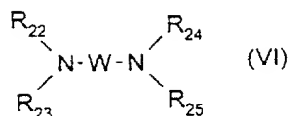
and aromatic alcohols such as benzyl alcohol or phenoxyethanol, similar products and mixtures thereof.

The solvents can be present in proportions preferably of between 1 and 40% by weight approximately
5 relative to the total weight of the ready-to-use dye composition, and even more preferably between 5 and 30% by weight approximately.

The pH of the ready-to-use composition in accordance with the invention is chosen such that the
10 enzymatic activity of the 2-electron oxidoreductase is sufficient. It is generally between 5 and 11 approximately, and preferably between 6.5 and 10 approximately. It can be adjusted to the desired value using acidifying or basifying agents usually used for
15 dyeing keratin fibres.

Among the acidifying agents, mention may be made, by way of example, of inorganic or organic acids such as hydrochloric acid, orthophosphoric acid, sulphuric acid, carboxylic acids such as acetic acid,
20 tartaric acid, citric acid or lactic acid, and sulphonic acids.

Among the basifying agents, mention may be made, by way of example, of aqueous ammonia, alkaline carbonates, alkanolamines such as mono-, di- and
25 triethanolamines, 2-methyl-2-aminopropanol and derivatives thereof, sodium hydroxide, potassium hydroxide and the compounds of formula (VI) below:



in which W is a propylene residue optionally substituted with a hydroxyl group or a C₁-C₄ alkyl radical; R₂₂, R₂₃, R₂₄ and R₂₅, which may be identical or different, represent a hydrogen atom or a C₁-C₄ alkyl or C₁-C₄ hydroxyalkyl radical.

The ready-to-use dye composition in accordance with the invention can also contain various adjuvants used conventionally in compositions for the dyeing of the hair, such as anionic, cationic, nonionic, amphoteric or zwitterionic surfactants or mixtures thereof, anionic, cationic, nonionic, amphoteric or zwitterionic polymers or mixtures thereof, inorganic or organic thickeners, antioxidants, enzymes other than the 2-electron oxidoreductases used in accordance with the invention, such as, for example, peroxidases, penetration agents, sequestering agents, fragrances, buffers, dispersing agents, conditioners such as, for example, silicones which may or may not be volatile or modified, film-forming agents, ceramides, preserving agents and opacifiers.

Needless to say, a person skilled in the art will take care to select this or these optional complementary compound(s) such that the advantageous properties intrinsically associated with the ready-to-use dye composition in accordance with the invention

are not, or are not substantially, adversely affected by the addition or additions envisaged.

The ready-to-use dye composition in accordance with the invention can be in various forms, such as in
5 the form of liquids, creams or gels, which may be pressurized, or in any other form which is suitable for dyeing keratin fibres, and in particular human hair. In this case the oxidation dyes and the 2-electron oxidoreductase(s) are present in the same ready-to-use
10 composition, and consequently the said composition must be free of gaseous oxygen, so as to avoid any premature oxidation of the oxidation dye(s).

A subject of the invention is also a process for dyeing keratin fibres, and in particular human
15 keratin fibres such as the hair, using the ready-to-use dye composition as defined above.

According to this process, at least one ready-to-use dye composition as defined above is applied to the fibres, for a period which is sufficient to develop
20 the desired coloration, after which the fibres are rinsed, optionally washed with shampoo, rinsed again and dried.

The time required to develop the coloration on the keratin fibres is usually between 3 and 60 minutes
25 and even more precisely between 5 and 40 minutes.

According to one specific embodiment of the invention, the process includes a preliminary step which consists in separately storing, on the one hand,

a composition (A) comprising, in a medium which is suitable for dyeing, at least one first oxidation base chosen from para-phenylenediamine derivatives, double bases, ortho-aminophenols and heterocyclic bases, at least one second oxidation base chosen from para-aminophenols, at least one meta-aminophenol as coupler, and, on the other hand, a composition (B) comprising, in a medium which is suitable for dyeing, at least one enzyme of 2-electron oxidoreductase type in the presence of at least one donor for the said enzyme, and then in mixing them together at the time of use, after which this mixture is applied to the keratin fibres.

Another subject of the invention is a multi-compartment dyeing device or "kit" or any other multi-compartment packaging system, a first compartment of which comprises composition (A) as defined above and a second compartment of which comprises composition (B) as defined above. These devices can be equipped with means for delivering the desired mixture onto the hair, such as the devices described in patent FR-2,586,913 in the name of the Applicant.

The examples which follow are intended to illustrate the invention without, however, limiting its scope.

The hair was dyed in the shades given in the

5 table below:

EXAMPLE	Shade obtained
1	Dark pearlescent blonde
2	Dark mahogany blonde

CLAIMS

1. Ready-to-use composition for the oxidation dyeing of keratin fibres, and in particular human keratin fibres such as the hair, characterized in that it comprises, in a medium which is suitable for dyeing:
- at least one first oxidation base chosen from para-phenylenediamine derivatives other than para-phenylenediamine, double bases, ortho-aminophenols and heterocyclic bases,
 - at least one second oxidation base chosen from para-aminophenols,
 - at least one meta-aminophenol as coupler,
 - at least one enzyme of 2-electron oxidoreductase type, and
 - at least one donor for the said enzyme.
2. Composition according to Claim 1, characterized in that the 2-electron oxidoreductase is chosen from pyranose oxidases, glucose oxidases, glycerol oxidases, lactate oxidases, pyruvate oxidases and uricases.
3. Composition according to Claim 1 or 2, characterized in that the 2-electron oxidoreductase is chosen from uricases of animal, microbiological or biotechnological origin.
4. Composition according to any one of the preceding claims, characterized in that the 2-electron oxidoreductase(s) represent(s) from 0.01 to 20% by

weight relative to the total weight of the ready-to-use dye composition.

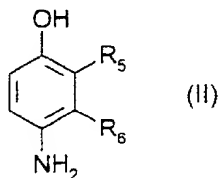
5. Composition according to Claim 4, characterized in that the 2-electron oxidoreductase(s) represent(s) from 0.1 to 5% by weight relative to the total weight of the ready-to-use dye composition.

6. Composition according to Claim 3, characterized in that the donor (or substrate) for the said 2-electron oxidoreductase is chosen from uric acid and its salts.

7. Composition according to any one of the preceding claims, characterized in that the donor(s) represent(s) from 0.01 to 20% by weight relative to the total weight of the ready-to-use dye composition.

8. Composition according to Claim 7, characterized in that the donor(s) represent(s) from 0.1 to 5% by weight relative to the total weight of the ready-to-use dye composition.

9. Composition according to any one of the preceding claims, characterized in that the para-aminophenols are chosen from the compounds corresponding to formula (II) below, and the addition salts thereof with an acid:



in which:

- R₅ represents a hydrogen or halogen atom or a C₁-C₄ alkyl, C₁-C₄ monohydroxyalkyl, (C₁-C₄)alkoxy(C₁-C₄)alkyl, C₁-C₄ aminoalkyl or hydroxy(C₁-C₄)alkylamino(C₁-C₄)alkyl radical,
 - 5 - R₆ represents a hydrogen or halogen atom or a C₁-C₄ alkyl, C₁-C₄ monohydroxyalkyl, C₂-C₄ polyhydroxyalkyl, C₁-C₄ aminoalkyl, cyano(C₁-C₄)alkyl or (C₁-C₄)alkoxy(C₁-C₄)alkyl radical,
- it being understood that at least one of the radicals R₅ or R₆ represents a hydrogen atom.

10. Composition according to Claim 9, characterized in that the para-aminophenols of formula (II) are chosen from para-aminophenol, 4-amino-3-methylphenol, 4-amino-3-fluorophenol, 4-amino-3-hydroxymethylphenol, 4-amino-2-methylphenol, 4-amino-2-hydroxymethylphenol, 4-amino-2-methoxymethylphenol, 4-amino-2-aminomethylphenol, 4-amino-2-(β -hydroxyethylaminomethyl)phenol and 4-amino-2-fluorophenol, and the addition salts thereof with an

20 acid.

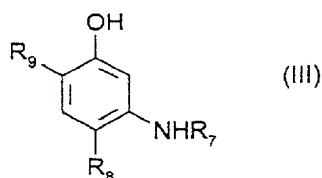
11. Composition according to any one of the preceding claims, characterized in that the para-aminophenol(s) represent(s) from 0.0005 to 12% by weight relative to the total weight of the ready-to-use

25 dye composition.

12. Composition according to Claim 11, characterized in that the para-aminophenol(s) represent(s) from 0.005 to 6% by weight relative to the

total weight of the ready-to-use dye composition.

13. Composition according to any one of the preceding claims, characterized in that the meta-aminophenols are chosen from the compounds of formula (III) below, and the addition salts thereof with an acid:



in which:

- R_7 represents a hydrogen atom or a C_1 - C_4 alkyl, C_1 - C_4 monohydroxyalkyl or C_2 - C_4 polyhydroxyalkyl radical,
- R_8 represents a hydrogen atom, a C_1 - C_4 alkyl or C_1 - C_4 alkoxy radical or a halogen atom chosen from chlorine, bromine and fluorine,
- R_9 represents a hydrogen atom or a C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 monohydroxyalkyl, C_2 - C_4 polyhydroxyalkyl, C_1 - C_4 monohydroxyalkoxy or C_2 - C_4 polyhydroxyalkoxy radical.

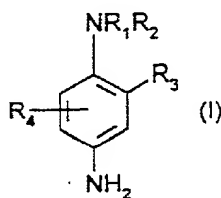
14. Composition according to Claim 13, characterized in that the meta-aminophenols of formula (III) are chosen from meta-aminophenol, 5-amino-2-methoxyphenol, 5-amino-3-(β -hydroxyethyloxy)phenol, 5-amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-2-methylphenol, 5-N-(β -hydroxyethyl)amino-4-methoxy-2-methylphenol, 5-amino-4-methoxy-2-methylphenol, 5-

amino-4-chloro-2-methylphenol, 5-amino-2,4-dimethoxyphenol and 5-(γ -hydroxypropylamino)-2-methylphenol and the addition salts thereof with an acid.

5 15. Composition according to any one of the preceding claims, characterised in that the meta-aminophenol(s) represent(s) from 0.0001 to 8% by weight relative to the total weight of the ready-to-use dye composition.

10 16. Composition according to Claim 15, characterized in that the meta-aminophenol(s) represent(s) from 0.005 to 5% by weight relative to the total weight of the dye composition.

15 17. Composition according to any one of the preceding claims, characterized in that the para-phenylenediamine derivatives are chosen from the compounds of formula (I) below, and the addition salts thereof with an acid:



in which:

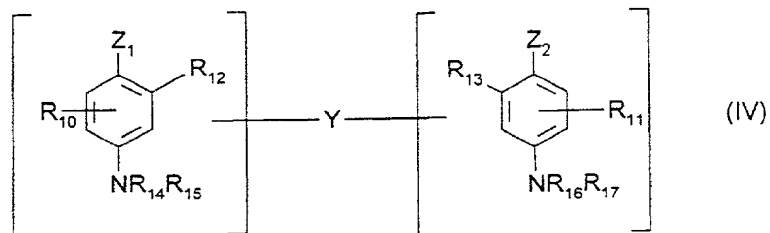
20 - R_1 represents a hydrogen atom, a C_1 - C_4 alkyl radical, a C_1 - C_4 monohydroxyalkyl radical, a C_2 - C_4 polyhydroxyalkyl radical, a $(\text{C}_1$ - C_4)alkoxy(C_1 - C_4)alkyl radical, a C_1 - C_4 alkyl radical substituted with a nitrogenous

- group, a phenyl radical or a 4'-aminophenyl radical;
- R_2 represents a hydrogen atom, a C_1-C_4 alkyl radical, a C_1-C_4 monohydroxyalkyl radical, a C_2-C_4 polyhydroxyalkyl radical, a (C_1-C_4) alkoxy (C_1-C_4) alkyl radical or a C_1-C_4 alkyl radical substituted with a nitrogenous group;
 - R_3 represents a hydrogen atom, a halogen atom such as a chlorine, bromine, iodine or fluorine atom, a C_1-C_4 alkyl radical, a C_1-C_4 monohydroxyalkyl radical, a C_1-C_4 hydroxyalkoxy radical, an acetylamino (C_1-C_4) -alkoxy radical, a C_1-C_4 mesylaminoalkoxy radical or a carbamoylamino (C_1-C_4) alkoxy radical,
 - R_4 represents a hydrogen or halogen atom or a C_1-C_4 alkyl radical;
- it being understood that at least one of the radicals R_1 to R_4 is other than a hydrogen atom.

18. Composition according to Claim 17, characterized in that the para-phenylenediamine derivatives of formula (I) are chosen from para-toluylenediamine, 2-chloro-para-phenylenediamine, 2,3-dimethyl-para-phenylenediamine, 2,6-dimethyl-para-phenylenediamine, 2,6-diethyl-para-phenylenediamine, 2,5-dimethyl-para-phenylenediamine, N,N-dimethyl-para-phenylenediamine, N,N-diethyl-para-phenylenediamine, N,N-dipropyl-para-phenylenediamine, 4-amino-N,N-diethyl-3-methylaniline, N,N-bis(β -hydroxyethyl)-para-phenylenediamine, 4-amino-N,N-bis(β -hydroxyethyl)-2-methylaniline, 4-amino-2-chloro-N,N-bis(β -hydroxy-

ethyl)aniline, 2- β -hydroxyethyl-para-phenylenediamine,
 2-fluoro-para-phenylenediamine, 2-isopropyl-para-
 phenylenediamine, N-(β -hydroxypropyl)-para-phenylene-
 diamine, 2-hydroxymethyl-para-phenylenediamine,
 5 N,N-dimethyl-3-methyl-para-phenylenediamine,
 N,N-(ethyl- β -hydroxyethyl)-para-phenylenediamine,
 N-(β,γ -dihydroxypropyl)-para-phenylenediamine,
 N-(4'-aminophenyl)-para-phenylenediamine, N-phenyl-
 para-phenylenediamine, 2- β -hydroxyethyloxy-para-
 10 phenylenediamine, 2- β -acetylaminioethyloxy-para-
 phenylenediamine and N-(β -methoxyethyl)-para-
 phenylenediamine, and the addition salts thereof with
 an acid.

19. Composition according to any one of the
 15 preceding claims, characterized in that the double
 bases are chosen from the compounds of formula (IV)
 below, and the addition salts thereof with an acid:



in which:

- Z_1 and Z_2 , which may be identical or different,
 20 represent a hydroxyl or $-\text{NH}_2$ radical which may be
 substituted with a C_1 - C_4 alkyl radical or with a linker
 arm Y;
- the linker arm Y represents a linear or branched

alkylene chain containing from 1 to 14 carbon atoms, which may be interrupted by or terminated with one or more nitrogenous groups and/or one or more hetero atoms such as oxygen, sulphur or nitrogen atoms, and

5 optionally substituted with one or more hydroxyl or C₁-C₆ alkoxy radicals;

- R₁₀ and R₁₁ represent a hydrogen or halogen atom, a C₁-C₄ alkyl radical, a C₁-C₄ monohydroxyalkyl radical, a C₂-C₄ polyhydroxyalkyl radical, a C₁-C₄ aminoalkyl
10 radical or a linker arm Y;

- R₁₂, R₁₃, R₁₄, R₁₅, R₁₆ and R₁₇, which may be identical or different, represent a hydrogen atom, a linker arm Y or a C₁-C₄ alkyl radical;

it being understood that the compounds of formula (IV)

15 contain only one linker arm Y per molecule.

20. Composition according to Claim 19, characterized in that the double bases of formula (IV) are chosen from N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-aminophenyl)-1,3-diaminopropanol, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4'-aminophenyl)ethylenediamine,
20 N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(β -hydroxyethyl)-N,N'-bis(4-aminophenyl)-tetramethylenediamine, N,N'-bis(4-methylaminophenyl)-tetramethylenediamine, N,N'-bis(ethyl)-N,N'-bis(4'-amino-3'-methylphenyl)ethylenediamine and
25 1,8-bis(2,5-diaminophenoxy)-3,5-dioxaoctane, and the addition salts thereof with an acid.

21. Composition according to any one of the

preceding claims, characterized in that the ortho-aminophenols are chosen from 2-aminophenol, 2-amino-5-methylphenol, 2-amino-6-methylphenol and 5-acetamido-2-aminophenol, and the addition salts thereof with an acid.

22. Composition according to any one of the preceding claims, characterized in that the heterocyclic bases are chosen from pyridine derivatives, pyrimidine derivatives, pyrazole derivatives and pyrazolopyrimidine derivatives, and the addition salts thereof with an acid.

23. Composition according to any one of the preceding claims, characterized in that the para-phenylenediamine derivative(s) and/or the double base(s) and/or the ortho-aminophenol(s) and/or the heterocyclic base(s) represent(s) from 0.0005 to 12% by weight relative to the total weight of the ready-to-use dye composition.

24. Composition according to Claim 23, characterized in that the para-phenylenediamine derivative(s) and/or the double base(s) and/or the ortho-aminophenol(s) and/or the heterocyclic base(s) represent(s) from 0.005 to 6% by weight relative to the total weight of the ready-to-use dye composition.

25. Composition according to any one of the preceding claims, characterized in that the addition salts with an acid are chosen from the hydrochlorides, hydrobromides, sulphates, tartrates, lactates and

acetates.

26. Composition according to any one of the preceding claims, characterized in that the medium which is suitable for dyeing consists of water or a mixture of water and at least one organic solvent.

27. Composition according to any one of the preceding claims, characterized in that it has a pH of between 5 and 11.

28. Composition according to any one of the preceding claims, characterized in that it contains at least one peroxidase.

29. Process for dyeing keratin fibres, and in particular human keratin fibres such as the hair, characterized in that at least one ready-to-use dye composition as defined in any one of the preceding claims is applied to the said fibres, for a period which is sufficient to develop the desired coloration.

30. Process according to Claim 29, characterized in that it includes a preliminary step which consists in separately storing, on the one hand, a composition (A) comprising, in a medium which is suitable for dyeing, at least one first oxidation base chosen from para-phenylenediamine derivatives, double bases, ortho-aminophenols and heterocyclic bases, at least one second oxidation base chosen from para-aminophenols, at least one meta-aminophenol as coupler, and, on the other hand, a composition (B) comprising, in a medium which is suitable for dyeing, at least one

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31. Multi-compartment dyeing device or "kit", characterized in that it includes a first compartment comprising composition (A) as defined in Claim 30 and a second compartment comprising composition (B) as defined in Claim 30.

ABSTRACT

COMPOSITION FOR THE OXIDATION DYEING OF KERATIN FIBRES AND DYEING PROCESS USING THIS COMPOSITION

The invention relates to a ready-to-use composition for the oxidation dyeing of keratin fibres, and in particular human keratin fibres such as the hair, comprising, in a medium which is suitable for dyeing, at least one first oxidation base chosen from para-phenylenediamine derivatives, double bases, ortho-aminophenols and heterocyclic bases, at least one second oxidation base chosen from para-aminophenols, at least one meta-aminophenol as coupler and at least one enzyme of 2-electron oxidoreductase type in the presence of at least one donor for the said enzyme, and to the dyeing process using this composition.

Declaration and Power of Attorney for Patent Application

Déclaration et Pouvoir pour Demand de Brevet

French Language Declaration

En tant que l'inventeur nommé ci-après, je déclare par le présent acte que:

Mon domicile, mon adresse postale et ma nationalité sont ceux figurant ci-dessous à côté de mon nom.

Je crois être le premier inventeur original et unique (si un seul nom est mentionné ci-dessous), ou l'un des premiers co-inventeurs originaux (si plusieurs noms sont mentionnés ci-dessous) de l'objet revendiqué, pour lequel une demande de brevet a été déposée concernant l'invention intitulée

et dont la description est fournie ci-joint à moins que la case suivante n'ait été cochée:

☒ a été déposée le _____
sous le numéro de demande des Etats-Unis ou le
numéro de demande international PCT
_____ et modifiée
_____ (les cas échéant).

Je déclare par le présent acte avoir passé en revue et compris le contenu de la description ci-dessus, revendications comprises, telles que modifiées par toute modification dont il aura été fait référence ci-dessus.

Je reconnais devoir divulguer toute information pertinente à la brevetabilité, comme défini dans le Titre 37, § 1.56 du Code fédéral des réglementations.

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

COMPOSITION FOR THE OXIDATION DYEING OF
KERATIN FIBRES AND DYEING PROCESS USING
THIS COMPOSITION

the specification of which is attached hereto unless the following box is checked:

☒ was filed on September 28, 1998 as United States Application Number or PCT International Application Number PCT/FR98/02074 and was amended on _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

French Language Declaration

Je revendique par le présent acte avoir la priorité étrangère, en vertu du Titre 35, § 119(a)-(d) ou § 365(b) du Code des Etats-Unis, sur toute demande étrangère de brevet ou certificat d'inventeur ou, en vertu du Titre 35, § 365(a) du même Code, sur toute demande internationale PCT désignant au moins un pays autre que les Etats-Unis et figurant ci-dessous et, en cochant la case, j'ai aussi indiqué ci-dessous toute demande étrangère de brevet, tout certificat d'inventeur ou toute demande internationale PCT ayant une date de dépôt précédant celle de la demande à propos de laquelle une priorité est revendiquée.

Prior foreign application(s)
Demande(s) de brevet antérieure(s)

<u>97/12351</u>	<u>France</u>
(Number)	(Country)
(Numéro)	(Pays)
<u> </u>	<u> </u>
(Number)	(Country)
(Numéro)	(Pays)

Je revendique par le présent acte tout bénéfice, en vertu du Titre 35, § 119(e) du Code des Etats-Unis, de toute demande de brevet provisoire effectuée aux Etats-Unis et figurant ci-dessous.

<u> </u>	<u> </u>
(Application No.)	(Filing Date)
(N° de demande)	(Date de dépôt)
<u> </u>	<u> </u>
(Application No.)	(Filing Date)
(N° de demande)	(Date de dépôt)

Je revendique par le présent acte tout bénéfice, en vertu du Titre 35, § 120 du Code des Etats-Unis, de toute demande de brevet effectuée aux Etats-Unis, ou en vertu du Titre 35, § 365(c) du même Code, de toute demande internationale PCT désignant les Etats-Unis et figurant ci-dessous et, dans la mesure où l'objet de chacune des revendications de cette demande de brevet n'est pas divulgué dans la demande antérieure américaine ou internationale PCT, en vertu des dispositions du premier paragraphe du Titre 35, § 112 du Code des Etats-Unis, je reconnais devoir divulguer toute information pertinente à la brevetabilité, comme défini dans le Titre 37, § 1.56 du Code fédéral des réglementations, dont laquelle est devenue disponible entre la date de dépôt de la demande antérieure, et la date de dépôt de la demande nationale ou internationale PCT de la présente demande:

<u> </u>	<u> </u>
(Application No.)	(Filing Date)
(N° de demande)	(Date de dépôt)
<u> </u>	<u> </u>
(Application No.)	(Filing Date)
(N° de demande)	(Date de dépôt)

I hereby claim foreign priority under Title 35, United States Code, § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International Application which designated at least one country other than the United States, listed below, and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Priority Not Claimed
Droit de priorité non revendiqué

<u>03 /October / 1997</u>	<input type="checkbox"/>
(Day/Month/Year Filed)	
(Jour/Mois/Anné de dépôt)	
<u> </u>	<input type="checkbox"/>
(Day/Month/Year Filed)	
(Jour/Mois/Anné de dépôt)	

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT International Application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International Application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose any or all information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

<u> </u>	(Status) (patented, pending, abandoned)
<u> </u>	(Status) (breveté, en cours d'examen, abandonné)
<u> </u>	(Status) (patented, pending, abandoned)
<u> </u>	(Status) (breveté, en cours d'examen, abandonné)

Je déclare par le présent acte que toute déclaration ci-incluse est, à ma connaissance, véridique et que toute déclaration formulée à partir de renseignements ou de suppositions est tenue pour véridique; et de plus, que toutes ces déclarations ont été formulées en sachant que toute fausse déclaration volontaire ou son équivalent est passible d'une amende ou d'une incarcération, ou des deux, en vertu de la Section 1001 du Titre 18 du Code des Etats-Unis, et que de telles déclarations volontairement fausses risquent de compromettre la validité de la demande de brevet ou du brevet délivré à partir de celle-ci.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

French Language Declaration

POUVOIRS: En tant que l'inventeur cité, je désigne par la présente l'(les) avocat(s) et/ou agent(s) suivant(s) pour qu'ils poursuive(nt) la procédure de cette demande de brevet et traite(nt) toute affaire s'y rapportant avec L'Office des brevets et des marques: (*mentionner le nom et le numéro d'enregistrement*).

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this patent application and transact all business in the Patent and Trademark Office connected therewith: (*list name and registration number*):

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Nom complet du deuxième co-inventeur, le cas échéant:	Full name of second joint inventor, if any: <u>Jean COTTERET</u>
Signature du deuxième inventeur Date	Second Inventor's signature Date <i>Jean COTTERET</i> May 19 th 1999
Domicile:	Residence <u>Verneuil-sur-Seine, France</u> <i>FRX</i>
Nationalité:	Citizenship French
Adresse postale:	Post Office Address 13, rue du Pré Rousselin, 78480 Verneuil-sur-Seine, France

Nom complet de l'unique ou premier inventeur:	Full name of sole or third inventor <u>Arnaud DE LABBEY</u>
Signature de l'inventeur Date	Third Inventor's signature Date <u>Arnaud de LABBEY</u> <u>05/21/1999</u>
Domicile	Residence <u>Aulnay Sous Bois, France</u> <u>FRX</u>
Nationalité:	Citizenship French
Adresse postale:	Post Office Address 9, rue Dordain, 93600 Aulnay Sous Bois, France
Nom complet du deuxième co-inventeur, le cas échéant:	Full name of fourth joint inventor, if any: <u>Mireille MAUBRU</u>
Signature du deuxième inventeur Date	Fourth Inventor's signature Date <u>Mireille MAUBRU</u> <u>05/20/99</u>
Domicile:	Residence <u>Chatou, France</u> <u>FRX</u>
Nationalité:	Citizenship French
Adresse postale:	Post Office Address 7, avenue d'Eprenesnil, 78400 Chatou, France

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4-00